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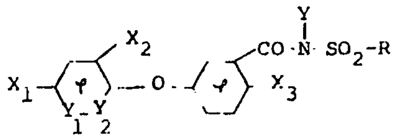
(54) Herbicidal N-sulphonyl-aryl oxybenzamide derivatives

(57) New herbicides especially useful against weeds in cereals or soya, have the formula:



(I)

in which: Y_1 is a nitrogen atom or a group $-CH=$, Y_2 is a nitrogen atom or, when Y_1 is a group $-CH=$, a group $-CX_4=$, X_1, X_2 and X_3, which are identical or different, each represent a halogen atom or a polyhalogenoalkyl, cyano, nitro, alkyl, alkoxy, alkylsulphonyl, $-SO_2NH_2$, nitroso or alkoxycarbonyl group, X_4 is as defined for X_1, X_2 and X_3 and can also represent hydrogen, R is a substituted or unsubstituted hydrocarbon radical or a substituted or unsubstituted heterocyclic ring and Y is a hydroxy group or a halogen atom.

GB 2 103 609 A

SPECIFICATION

Herbicidal phenoxybenzoic acid derivatives

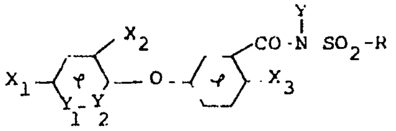
The present invention relates to herbicidal compounds, to compositions containing them and to their use in controlling the growth of weeds.

Herbicides are known which are derived from phenoxybenzoic acid and more particularly from 5-[2-chloro-4-(trifluoromethyl)-phenoxy]-2-nitrobenzoic acid, known under the name acifluorfen.

Various derivatives of these compounds have already been proposed, including the alkyl, cycloalkyl, thioalkyl and phenyl esters, the monoalkylaminedes or dialkylamides and the acid chlorides. Amongst the U.S. Patents describing such compounds, there may be mentioned U.S. Patents 3,652,645, 3,784,635, 3,873,302, 3,983,168, 3,907,866, 3,798,276, 3,928,416 and 4,063,929.

Despite the existence of a large number of derivatives of phenoxybenzoic acids, or even of simple diphenyl ethers, it is desirable to have available further herbicides of these families, so as to be able to solve various possible problems of desirable herbicidal treatments, especially pre-emergence treatments.

The present invention provides herbicidal compounds of the formula:



(I)

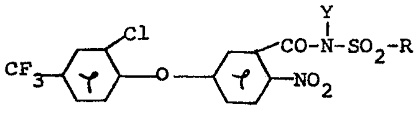
in which: Y_1 is a nitrogen atom or a group $-CH=$, Y_2 is a nitrogen atom or, when Y_1 is a group $-CH=$, a group $-CX_4=$, X_1, X_2 and X_3, which are identical or different, each represent a halogen atom (in particular chlorine, bromine or fluorine), a polyhalogenoalkyl group, in particular a trifluoromethyl group, a cyano or nitro group or an alkyl, alkoxy, alkylsulphonyl, SO_2NH_2, nitroso or alkoxycarbonyl group, X_4 is as defined for X_1, X_2 and X_3 and can also represent hydrogen, R is a substituted or unsubstituted hydrocarbon radical preferably having from 1 to 12 carbon atoms, in particular a substituted or unsubstituted alkyl or aryl radical, or a substituted or unsubstituted hetero-cyclic ring having from 5 to 7 atoms in the ring, and Y is a hydroxy group or, preferably, a halogen atom, especially chlorine. Alkyl groups and moieties within the definition of X_1 to X_4 preferably contain from 1 to 4 carbon atoms.

Preferred compounds of formula I are those in which alkyl groups and moieties within the definition of X_1 to X_4 contain from 1 to 4 carbon atoms, and in which R is a substituted or unsubstituted hydrocarbon radical having from 1 to 12 carbon atoms or a substituted or unsubstituted heterocyclic ring having from 5 to 7 atoms in the ring.

Preferred radicals R which may be mentioned are alkyl radicals having 1 to 4 carbon atoms; aryl radicals, especially the phenyl radical, which are unsubstituted or substituted by 1 to 4 alkyl radicals each having from 1 to 4 carbon atoms, by chlorine or bromine atoms, by hydroxy, SH, CN or COOH groups or by alkoxy, thioalkyl or alkoxy-carbonyl groups, the alkyl parts of these groups having from 1 to 4 carbon atoms; and alkenyl and alkynyl groups having from 2 to 6 carbon atoms.

Amongst the compounds of the formula (I), a very particularly advantageous class is represented by the compounds in which X_1 and X_2 are polyhalogenoalkyl groups (preferably CF_3) or halogen atoms, Y_1 and Y_2 both represent the group $-CH=$ and X_3 is the nitro group of a halogen atom. The compounds in which X_1 is Cl or CF_3, X_2 is Cl, X_3 is Cl or NO_2, and Y_1 and Y_2 are both $-CH=$ are more particularly preferred.

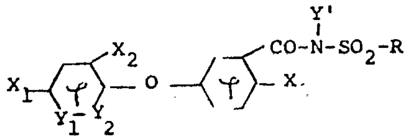
Compounds of the formula:



(II)

wherein Y and R are as hereinbefore defined are particularly preferred: amongst these compounds, preference is also given to those in which R is an alkyl group (preferably of 1 to 4 carbon atoms) and Y is a chlorine atom.

In general, the compounds according to the invention can be prepared from precursors of the formula:



(III)

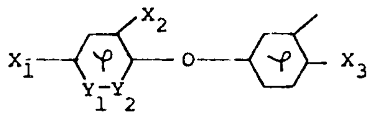
in which Y′ is the hydrogen atom or an alkali metal atom, preferably sodium, the other symbols being as hereinbefore defined.

The majority of the compounds of the formula (III), or their direct homologues, have already been described in European Patent Application Nos. 3,416 or 23,392: compounds or formula III which are not specifically described therein may be prepared by the application or adaptation of known methods.

The compounds according to the invention in which Y is a halogen atom are preferably prepared by reacting a hypohalite, preferably in alkali metal, e.g. sodium, hypohalite, with a precursor of the formula (III) in which Y′ is a hydrogen atom.

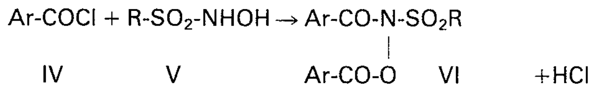
The reaction is generally carried out in aqueous or aqueous-organic solution or suspension, in the presence of a water-miscible solvent, such as an alcohol or a ketone, in particular methanol, ethanol, acetone, methyl ethyl ketone or methyl isobutyl ketone. The reaction is generally carried out in an aqueous medium at from −10 to +60°C, preferably from 10 to 40°C, the concentration of reactant of the formula (III) and reaction product of the formula (I) according to the invention, in the reaction medium, generally being from 0.5 to 30% by weight, preferably from 2 to 10%; the amount of hypohalite used is generally from 1 to 3 times the stoichiometric amount; the pH during the reaction is generally more than 10, preferably more than 11.5.

The compounds according to the invention in which Y is a hydroxy group may be prepared by reacting a compound of formula IV with a compound of formula V to obtain a compound of formula VI and then reacting the compound of formula VI with an alkali metal alkoxide or with a solution of an alkali metal hydroxide in an alcohol R^1OH in which R^1 represents an alkyl group and then with a strong acid to obtain the compound of formula I in which Y is a hydroxy group, in accordance with the following reaction scheme, in which the symbol Ar represents the radical:



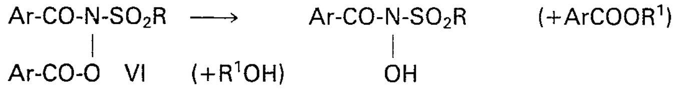
R^1 represents an alkyl group, preferably containing 1 to 4 carbon atoms and the other symbols are as hereinbefore defined:

Reaction scheme



$$\underset{\mathrm{IV}}{\mathrm{Ar\text{-}COCl}} + \underset{\mathrm{V}}{\mathrm{R\text{-}SO_2\text{-}NHOH}} \rightarrow \begin{array}{l}\mathrm{Ar\text{-}CO\text{-}N\text{-}SO_2R}\\ \qquad\qquad\ \ |\\ \mathrm{Ar\text{-}CO\text{-}O}\quad \mathrm{VI}\end{array} \quad +\mathrm{HCl}$$

then



$$\begin{array}{l}\mathrm{Ar\text{-}CO\text{-}N\text{-}SO_2R}\\ \qquad\qquad\ \ |\\ \mathrm{Ar\text{-}CO\text{-}O}\quad \mathrm{VI}\end{array} \xrightarrow{} \ (+R^1OH) \quad \begin{array}{l}\mathrm{Ar\text{-}CO\text{-}N\text{-}SO_2R}\\ \qquad\qquad\ \ |\\ \qquad\qquad\ \mathrm{OH}\end{array} \quad (+\mathrm{ArCOOR^1})$$

The first of these reactions is advantageously carried out in the presence of an acid acceptor, such as a tertiary amine, in particular pyridine.

The second of these reactions, which is an alcoholysis, is carried out by reaction with an alkali metal alkoxide (or a solution of an alkali metal hydroxide in an alcohol designated by R^1OH in the scheme) and then by reaction with a strong acid, such as hydrochloric acid.

The compounds according to the invention are particularly advantageous both for their selectivity and for their herbicidal activity in pre-emergence and post-emergence treatments. Amongst the crops which can be treated with the products of the invention, there may be mentioned soya, cotton, rice, groundnut and cereals, including maize.

By the expression "known methods" as used in this specification is meant methods heretofore used or described in the chemical literature.

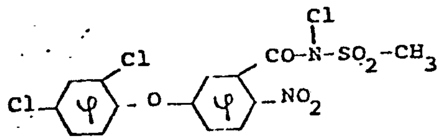
Unless otherwise indicated in this specification percentages are by weight.

The following Examples illustrate the invention and show how it can be put into practice.

Example 1:

5-(2,4-Dichlorophenoxy)-2-nitro-N-methane-sulphonylbenzamide (m.p.: 181°C) (0.5 g) is dissolved in an aqueous solution of sodium hypochlorite (20 cc) containing 1.5 mols/l of hypochlorite and containing sodium hydroxide to bring the pH to 12. The solution is stirred for 30 minutes at 20°C. Acetic acid is added dropwise until a pH of 7.5 is reached; the stirring is then continued for a further 30 minutes.

The precipitate is filtered off, washed with cold water and dried in air; this gives 5-(2,4-dichlorophenoxy)-2-nitro-N-chloro-N-methanesulphonylbenzamide (0.3 g) having an m.p. of 161-163°C and the Formula:

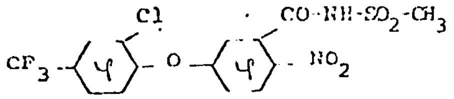


This compound has an infra-red absorption band at 1,728 cm^{-1} (carbonyl band), whereas the corresponding band for the starting reactant is at 1,688 cm^{-1}.

Example 2:

Javelle water (4.46 1), containing 1.57 mols/l of sodium hypochlorite and 105 ml of 30% strength by weight NaOH solution to bring the pH to 12.2, is added to water (10 litres).

5-[2-Chloro-4-(trifluoromethyl)-phenoxy]-2-nitro-N-methanesulphonylbenzamide (438 g), having an m.p. of 215-218°C and the formula:

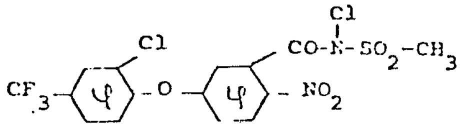


is added gradually at 18-20°C, with stirring, over a period of 1 hour 45 minutes.

The stirring is continued for 4 hours.

The mixture is then neutralised by adding acetic acid (215 ml) gradually over a period of 20 minutes, the temperature being kept at 18-20°C.

The white precipitate is filtered off, washed with water, drained and dried in vacuo in the presence of P_2O_5. This gives 5-[2-chloro-4-(trifluoromethyl)-phenoxy]-2-nitro-N-chloro-N-methanesulphonylbenzamide (279 g) having an m.p. of 161-162°C. It can be recrystallised from a diethyl ether/chloroform mixture and has the formula:



In the nuclear magnetic resonance spectrum, it does not show an NH band, and in the infra-red spectrum, it shows the carbonyl absorption band at 1,728 cm^{-1}.

Example 3:

Species of crops and weeks are placed in 10 cm row in 20 cm × 25 cm dishes containing earth.

The species used are indicated in Table (I).

Cotton, maize (field corn), soya and xanthium are sown at a rate of 4 to 5 seeds per row.

The smaller species (abutilon, wild mustard, amaranth, foxtail millet and green panic grass (Setaria viridis)) are sown without counting the seeds, but in a number which is nevertheless sufficient subsequently to form a dense line of plantlets.

The initial watering, up to the emergence period, is carried out from above the earth covering the seeds.

The pre-emergence treatment is carried out less than one day after sowing.

The desired stage of development for the post-emergence treatment of cotton, soya, xanthium, abutilon, wild mustard and chenopodium is the stage of one true leaf or a first trifoliate leaf. For maize, the desired stage is a height of 7.5 to 10 cm, whereas for the graminaceous plants, the desired stage is a height of 2.5 cm.

The compositions of the invention are applied by spraying under a pressure of 2.5 bars (= 36 psi) at a rate of 375 1/ha (= 40 gallons/acre). The spraying compositions consisted of a mixture, in respective proportions by volume, of water (20 cc) and 0.1% of a surface-active agent (a mixture of alkali metal salts of condensates of ethylene oxide with alkyl (C_6-C_{16})-benzene-sulphonic acids).

After treatment, irrigation is carried out by sub-irrigation in the case of the plantlets which have already emerged, and by watering from above in the case of the seeds which have not yet produced plantlets.

2 weeks after treatment, the activity measurements are carried out according to a scale from 0 to 100%. In the case of the weeds, this measurement indicates the level of destruction; in the case of the crops, this measurement indicates the degree of attack or degree of phytotoxicity, the grade zero being assigned to plants in the same condition as the conrol, and the grade one hundred being assigned to complete destruction.

As regards the application doses, the correspondance between metric and U.S. units is as follows:

pounds/acre	10	4	2	1	0.5	0.25	0.125	0.0625
kg/ha	11.2	4.48	2.24	1.12	0.56	0.28	0.14	0.07

Table (II) gives the results obtained with the compounds of Examples 1 and 2.

Example 4: Herbicidal application in the pre-emergence treatment of plant species.

A number of seeds are sown in 9 × 9× 9 cm pots filled with light agricultural earth, this number being determined as a function of the plant species and the size of the seed.

The seeds are then covered with an approximately 3 mm thick layer of earth.

After moistening the earth, the pots are treated by spraying with an amount of spraying mixture which corresponds to a volumetric application dose of 500 1/ha and contains the active ingredient at the desired concentration.

The spraying mixture was prepared by diluting a wettable powder, which has itself prepared from:
active ingredient (500 g)
sodium alkylnaphthalenesulphonate (15 g)
methylene-bis-(sodium naphthalenesulphonate) (50 g)
silica (50 g)
kaolin (390 g)

According to the concentration of active ingredient in the spraying mixture, the dose of active ingredient applied was 0.25 to 1 kg/ha.

The treated pots are then placed in troughs which are intended to receive the moistening water, by sub-irrigation, and are kept for 21 days at a temperature of 22-24°C and under 70% relative humidity.

After 21 days, the number of living plants in the pots treated with the spraying mixture containing the active ingredient to be tested, and the number of living plants in a control pot treated under the same conditions, but by means of a spraying mixture not containing active ingredient, are counted. The percentage destruction of the treated plants, relative to the untreated control, is thus determined. A percentage destruction equal to 100% indicates that there has been complete destruction of the plant species in question, and a percentage of 0% indicates that the number of living plants in the treated pot is identical to that in the control pot.

Example 5: Herbicidal application in the post-emergence treatment of plant species

A number of seeds are sown in 9 × 9 × 9 cm pots filled with light agricultural earth, this number being determined as a function of the plant species and the size of the seed.

The seeds are then covered with an approximately 3 mm thick layer of earth and the seed is left to germinate until it has produced a plantlet of 5 to 10 cm in height.

The pots are then treated by spraying with an amount of spraying mixture which corresponds to a volumetric application does of 500 1/ha and contains the active ingredient at the desired concentration.

The spraying mixture was prepared in the same manner as in Example 4.

According to the concentration of active ingredient in the spraying mixture, the dose of active ingredient applied was 0.125 to 1 kg/ha.

The treated pots are then placed in troughs which are intended to receive the moistening water, by sub-irrigation, and are kept for 14 days at a temperature of 22-24°C and under 70% relative humidity.

After 14 days, the number of living plants in the pots treated with the spraying mixture containing the active ingedient to be tested, and the number of living plants in a control pot treated under the same conditions, but by means of a spraying mixture not containing active ingredient, are counted. The percentage destruction of the treated plants, relative to the untreated control, is thus determined. A percentage destruction equal to 100% indicates that there has been complete destruction of the plant species in question, and a percentage of 0% indicates that the number of living plants in the treated pot is identical to that in the control pot.

The results of Examples 4 and 5 are indicated in Table (III).

This Table (III) comprises, on the one hand, pre-emergence results obtained in accordance with Example 4, and, on the other hand, post-emergence results obtained in accordance with Example 5.

The experiments carried out therefore show the notably advantageous properties of the compounds according to the invention, both for the pre-emergence treatment of crops, more particularly of soya and cereals, and for the post-emergence treatment, in particular of soya and cereals. In the case of soya, the activity of the compounds is particularly advantageous if this crop is infested with dicotyledon weeds such as abutilon, xanthium and ipomea. In the case of cereals, the activity of the compounds is particularly advantageous if this crop is infested with dicotyledon weeds, in particular polygonum, amaranth, ambrosia, chrysanthemum, stellaria and galium.

For their use in practice, the compounds according to the invention are rarely employed by themselves. Most frequently, the compounds form part of compositions. The present invention provides herbicidal compositions which comprise, as active ingredient, a compound of formula (I) in association with a herbicidally acceptable carrier. The carrier may be solid or liquid. The compositions may also comprise herbicidally acceptable surface-active agents. In particular, the customary inert carriers and the customary surface-active agents can be used.

These compositions can also contain various other ingredients, such as e.g. protective colloids, adhesives, thickeners, thixotropic agents, penetrating agents, stabilisers and sequestering agents, and also other known active ingredients having pesticidal properties (in particular insecticidal, fungicidal or herbicidal properties), properties promoting plant growth (in particular fertilisers) or properties regulating plant growth. More generally, the compounds used in the invention can be combined with any of the solid or liquid additives corresponding to the usual formulation techniques.

The use doses of the compounds of the invention can vary within wide limits, in particular according to the nature of the adventitious plants to be removed and the usual degree of infestation of the crops by these adventitious plants.

In general, the compositions according to the invention usually contain from 0.05 to about 95% (by weight) of active ingredient: such compositions generally contain from 1% to about 95% of one or more solid or liquid carriers and, if appropriate, from 0.1 to about 20% of one or more surface-active agents. Preferred solid compositions contain from 20 to 95% of active ingredient.

According to what has already been stated, the compounds used in the invention are generally combined with carriers and, if appropriate, surface-active agents.

In the present account, the term "carrier" denotes an organic or inorganic, natural or synthetic material with which the active ingredient is combined in order to facilitate its application to the plant, or to the soil. This carrier is therefore generally inert and it must be acceptable in agriculture, in particular on the plant treated. The carrier can be solid (e.g. clays, natural or synthetic silicates, silica, resins, waxes and solid fertilisers) or liquid (water, alcohols, in particular butanol, esters, in particular methylglycol acetate, ketones, in particular cyclohexanone and isophorone, petroleum fractions, aromatic hydrocarbons, in particular xylenes, or paraffinic hydrocarbons, aliphatic chlorohydrocarbons, in particular trichloroethane, or aromatic chlorohydrocarbons, in particular chlorobenzenes, water-soluble solvents, such as dimethylformamide, dimethyl sulphoxide and N-methylpyrrolidone, and liquefied gases.

The surface-active agent can be an emulsifying, dispersing or wetting agent of ionic or non-ionic type or a mixture of such surface-active agents. Examples which may be mentioned are salts of polyacrylic acids, salts of lignosulphonic acids, salts of phenolsulphonic or naphthalenesulphonic acids, polycondensates of ethylene oxide with fatty alcohols, fatty acids or fatty amines, substituted phenols (in particular alkylphenols or arylphenols), salts of sulphosuccinic acid esters, taurine derivatives (in particular alkyltaurates), phosphoric acid esters of condensates of ethylene oxide with alcohols or phenols, fatty acid esters of polyols, and derivatives of the above compounds containing sulphate, sulphonate and phosphate groups. The presence of at least one surface-active agent is generally essential if the active ingredient and/or the inert carrier are not soluble in water and if the vehicle of application is water.

The following Examples illustrate herbicidal compositions according to the invention.

As forms of solid compositions, there may be mentioned dusting powders (with a content of compound of the formula (I) which can range up to 100%) and granules, in particular those obtained by extrusion, by compaction, by the impregnation of a granular carrier or by the formation of granules from a powder (the content of compound of the formula (I) in the granules being between 0.5 and 80%). The solid compositions most frequently contain 20 to 80% of active ingredient.

As forms of liquid compositions or compositions which are to be made up into liquid compositions for application, there may be mentioned solutions, in particular emulsifiable concentrates, emulsions, suspension concentrates, aerosols, wettable powders (or spraying powders), dry flowable and pastes.

The liquid compositions most frequently contain 10 to 80% of active ingredient.

The emulsifiable or soluble concentrates most frequently comprise 10 to 80% of active ingredient, and the emulsions or solutions which are ready for application contain 0.01 to 20% of active ingredient. In addition to the solvent, the emulsifiable concentrates can contain, where necessary, 2 to 20% of suitable additives, such as stabilisers, surface-active agents, penetrating agents, corrosion inhibitors, dyestuffs and adhesives.

Starting from these concentrates, emulsions or solutions of any desired concentration, which are particularly suitable for application to the plants, can be obtained by dilution with water.

Examples of emulsifiable concentrates are now given:

Example 4

active ingredient	250 g
ethylene oxide/alkylphenol condensate	30 g
calcium alkylarylsulphonate	50 g
petroleum distillation cut distilling at between 160 and 185°C	670 g

Another formulation is:

Example 5

active ingredient	350 g
ethylene oxide/castor oil condensate	60 g
sodium alkylarylsulphonate	40 g
cyclohexanone	150 g
xylene	400 g

Another formulation is:

Example 6

active ingredient	400 g
ethylene oxide/alkylphenol condensate	100 g
ethylene glycol methyl ether	250 g
aromatic petroleum cut distilling at between 160 and 185°C	250 g

Another formulation is:

Example 7

active ingredient	400 g
phosphate of ethylene oxide/tristyrylphenol condensate	50 g
phosphate of ethylene oxide/alkylphenol condensate	65 g
sodium alkylbenzenesulphonate	35 g
cyclohexanone	300 g
aromatic petroleum cut distilling at between 160 and 185°C	150 g

Another formulation is:

Example 8

active ingredient	400 g/l
alkali metal dodecylbenzenesulphonate	24 g/l
10:1 ethylene oxide/nonylphenol condensate	16 g/l
cyclohexanone	200 g/l
aromatic solvent	q.s.p. 1 litre

Another formulation of an emulsifiable concentrate uses the following:

Example 9

active ingredient	250 g
epoxidised vegetable oil	25 g
mixture of an alkylarylsulphonate and a polyglycol ether of fatty alcohols	100 g
dimethylformamide	50 g
xylene	575 g

The suspension concentrates, which can be applied by spraying, are prepared so as to give a stable fluid product which does not form a deposit (by fine grinding), and they usually contain from 10 to 75% of active ingredient, from 0.5 to 15% of surface-active agents, from 0.1 to 10% of thixotropic agents, from 0 to 10% of suitable additives, such as anti-foam agents, corrosion inhibitors, stabilisers, penetrating agents and adhesives, and, as the carrier, water or an organic liquid in which the active ingredient is sparingly soluble or insoluble; certain organic solids, or inorganic salts, can be dissolved in the carrier in order to assist in preventing sedimentation or to act as anti-freeze agents for the water.

herbicidally acceptable carrier. The carrier may be solid or liquid. The compositions may also comprise herbicidally acceptable surface-active agents. In particular, the customary inert carriers and the customary surface-active agents can be used.

These compositions can also contain various other ingredients, such as e.g. protective colloids, adhesives, thickeners, thixotropic agents, penetrating agents, stabilisers and sequestering agents, and also other known active ingredients having pesticidal properties (in particular insecticidal, fungicidal or herbicidal properties), properties promoting plant growth (in particular fertilisers) or properties regulating plant growth. More generally, the compounds used in the invention can be combined with any of the solid or liquid additives corresponding to the usual formulation techniques.

The use doses of the compounds of the invention can vary within wide limits, in particular according to the nature of the adventitious plants to be removed and the usual degree of infestation of the crops by these adventitious plants.

In general, the compositions according to the invention usually contain from 0.05 to about 95% (by weight) of active ingredient: such compositions generally contain from 1% to about 95% of one or more solid or liquid carriers and, if appropriate, from 0.1 to about 20% of one or more surface-active agents. Preferred solid compositions contain from 20 to 95% of active ingredient.

According to what has already been stated, the compounds used in the invention are generally combined with carriers and, if appropriate, surface-active agents.

In the present account, the term "carrier" denotes an organic or inorganic, natural or synthetic material with which the active ingredient is combined in order to facilitate its application to the plant, or to the soil. This carrier is therefore generally inert and it must be acceptable in agriculture, in particular on the plant treated. The carrier can be solid (e.g. clays, natural or synthetic silicates, silica, resins, waxes and solid fertilisers) or liquid (water, alcohols, in particular butanol, esters, in particular methylglycol acetate, ketones, in particular cyclohexanone and isophorone, petroleum fractions, aromatic hydrocarbons, in particular xylenes, or paraffinic hydrocarbons, aliphatic chlorohydrocarbons, in particular trichloroethane, or aromatic chlorohydrocarbons, in particular chlorobenzenes, water-soluble solvents, such as dimethylformamide, dimethyl sulphoxide and N-methylpyrrolidone, and liquefied gases.

The surface-active agent can be an emulsifying, dispersing or wetting agent of ionic or non-ionic type or a mixture of such surface-active agents. Examples which may be mentioned are salts of polyacrylic acids, salts of lignosulphonic acids, salts of phenolsulphonic or naphthalenesulphonic acids, polycondensates of ethylene oxide with fatty alcohols, fatty acids or fatty amines, substituted phenols (in particular alkylphenols or arylphenols), salts of sulphosuccinic acid esters, taurine derivatives (in particular alkyltaurates), phosphoric acid esters of condensates of ethylene oxide with alcohols or phenols, fatty acid esters of polyols, and derivatives of the above compounds containing sulphate, sulphonate and phosphate groups. The presence of at least one surface-active agent is generally essential if the active ingredient and/or the inert carrier are not soluble in water and if the vehicle of application is water.

The following Examples illustrate herbicidal compositions according to the invention.

As forms of solid compositions, there may be mentioned dusting powders (with a content of compound of the formula (I) which can range up to 100%) and granules, in particular those obtained by extrusion, by compaction, by the impregnation of a granular carrier or by the formation of granules from a powder (the content of compound of the formula (I) in the granules being between 0.5 and 80%). The solid compositions most frequently contain 20 to 80% of active ingredient.

As forms of liquid compositions or compositions which are to be made up into liquid compositions for application, there may be mentioned solutions, in particular emulsifiable concentrates, emulsions, suspension concentrates, aerosols, wettable powders (or spraying powders), dry flowable and pastes.

The liquid compositions most frequently contain 10 to 80% of active ingredient.

The emulsifiable or soluble concentrates most frequently comprise 10 to 80% of active ingredient, and the emulsions or solutions which are ready for application contain 0.01 to 20% of active ingredient. In addition to the solvent, the emulsifiable concentrates can contain, where necessary, 2 to 20% of suitable additives, such as stabilisers, surface-active agents, penetrating agents, corrosion inhibitors, dyestuffs and adhesives.

Starting from these concentrates, emulsions or solutions of any desired concentration, which are particularly suitable for application to the plants, can be obtained by dilution with water.

Examples of emulsifiable concentrates are now given:

Example 4

active ingredient	250 g
ethylene oxide/alkylphenol condensate	30 g
calcium alkylarylsulphonate	50 g
petroleum distillation cut distilling at between 160 and 185°C	670 g

Another formulation is:

Example 5

active ingredient	350 g
ethylene oxide/castor oil condensate	60 g
sodium alkylarylsulphonate	40 g
cyclohexanone	150 g
xylene	400 g

Another formulation is:

Example 6

active ingredient	400 g
ethylene oxide/alkylphenol condensate	100 g
ethylene glycol methyl ether	250 g
aromatic petroleum cut distilling at between 160 and 185°C	250 g

Another formulation is:

Example 7

active ingredient	400 g
phosphate of ethylene oxide/tristyrylphenol condensate	50 g
phosphate of ethylene oxide/alkylphenol condensate	65 g
sodium alkylbenzenesulphonate	35 g
cyclohexanone	300 g
aromatic petroleum cut distilling at between 160 and 185°C	150 g

Another formulation is:

Example 8

active ingredient	400 g/l
alkali metal dodecylbenzenesulphonate	24 g/l
10:1 ethylene oxide/nonylphenol condensate	16 g/l
cyclohexanone	200 g/l
aromatic solvent	q.s.p. 1 litre

Another formulation of an emulsifiable concentrate uses the following:

Example 9

active ingredient	250 g
epoxidised vegetable oil	25 g
mixture of an alkylarylsulphonate and a polyglycol ether of fatty alcohols	100 g
dimethylformamide	50 g
xylene	575 g

The suspension concentrates, which can be applied by spraying, are prepared so as to give a stable fluid product which does not form a deposit (by fine grinding), and they usually contain from 10 to 75% of active ingredient, from 0.5 to 15% of surface-active agents, from 0.1 to 10% of thixotropic agents, from 0 to 10% of suitable additives, such as anti-foam agents, corrosion inhibitors, stabilisers, penetrating agents and adhesives, and, as the carrier, water or an organic liquid in which the active ingredient is sparingly soluble or insoluble; certain organic solids, or inorganic salts, can be dissolved in the carrier in order to assist in preventing sedimentation or to act as anti-freeze agents for the water.

A composition of a suspension concentrate is now given as an example:

Example 10

active ingredient	50 g
phosphate of ethylene oxide/tristyrylphenol condensate	50 g
ethylene oxide/alkylphenol condensate	50 g
sodium polycarboxylate	20 g
ethylene glycol	50 g
organopolysiloxane oil (anti-foam agent)	1 g
polysaccharide	12.5 g
water	316.5 g

The wettable powders (or spraying powders) are usually prepared so as to contain 20 to 95% of active ingredient, and they usually contain, in addition to a solid carrier, from 0 to 5% of a wetting agent, from 3 to 10% of a dispersing agent and, where necessary, from 0 to 10% of one or more stabilisers and/or other additives, such as penetrating agents, adhesives, anti-caking agents and dyestuffs.

Various compositions of wettable powders are now given as examples:

Example 11

active ingredient	50 %
calcium lignosulphonate (deflocculant)	5 %
isopropylnaphthalenesulphonate (anionic wetting agent)	1 %
anti-caking silica	5 %
kaolin (filler)	39 %

Another example of a wettable powder, this time of 80% strength, is given below:

Example 12

active ingredient	80 %
sodium alkylnaphthalenesulphonate	2%
sodium lignosulphonate	2%
anti-caking silica	3 %
kaolin	13 %

Another example of a wettable powder is given below:

Example 13

active ingredient	50 %
sodium alkylnaphthalenesulphonate	2 %
low-viscosity methylcellulose	2 %
diatomaceous earth	46 %

Another example of a wettable powder is given below:

Example 14

active ingredient	90 %
sodium dioctyl-sulphosuccinate	0.2 %
synthetic silica	9.8 %

Another composition of a spraying powder, this time of 40% strength, uses the following constituents:

Example 15

active ingredient	400 g
sodium lignosulphonate	50 g
sodium dibutylnaphthalenesulphonate	10 g
silica	540 g

Another composition of a spraying powder, this time of 25% strength, uses the following constituents:

Example 16

active ingredient	250 g
isooctylphenoxy-polyoxyethylene-ethanol	25 g
mixture of equal parts by weight of Champagne chalk and hydroxyethylcellulose	17 g
sodium aluminosilicate	543 g
kieselguhr	165 g

Another composition of a spraying powder, this time of 10% strength, uses the following constituents:

Example 17

active ingredient	100 g
mixture of sodium salts of saturated fatty acid sulphates	30 g
naphthalenesulphonic acid/formaldehyde condensate	50 g
kaolin	820 g

To obtain these spraying powders or wettable powders, the active ingredients are intimately mixed with the additional substances, in suitable mixers, or a porous filler is impregnated with the molten active ingredient, and the mixture is ground in mills or other suitable grinders. This gives spraying powders of advantageous wettability and suspendability; they can be suspended in water at any desired concentration, and this suspension can be used very advantageously, in particular for application to the leaves of plants.

The dry flowables (more exactly, these are granules which are readily dispersible in water) have a composition substantially similar to that of the wettable powders. They can be prepared by the formation of granules from formulations described for the wettable powders, either by a wet process (bringing the finely divided active ingredient into contact with the inert filler and with a small amount of water, e.g. 1 to 20%, or of an aqueous solution of dispersing agent or binder, followed by drying and sieving) or by a dry process (compaction, followed by grinding and sieving).

A formulation of a dry flowable is now given as an example:

Example 18

active ingredient	800 g
sodium alkylnaphthalenesulphonate	20 g
methylene-bis-(sodium naphthalene sulphonate)	80 g
kaolin	100 g

In place of the wettable powders, it is possible to produce pastes. The conditions and modes of production and use of these pastes are similar to those of the wettable powders or spraying powders.

As already stated, the aqueous dispersions and emulsions, e.g. compositions obtained by diluting, with water, a wettable powder or an emulsifiable concentrate according to the invention, are included within the general scope of the compositions which can be used in the present invention. The emulsions can be of the water-in-oil or oil-in-water type and they can have a thick consistency such as that of a "mayonnaise".

All these aqueous dispersions or emulsions, or spraying mixtures, can be applied to the crops in which weeds are to be destroyed, by any suitable method, mainly by spraying, at doses which are generally of the order of 100 to 1,200 litres of spraying mixture per hectare.

The granules, which are intended to be placed on the soil, are usually prepared so that they have dimensions of between 0.1 and 2 mm, and they can be manufactured by agglomeration or impregnation. Preferably, the granules contain 1 to 25% of active ingredient and 0 to 10% of additives, such as stabilisers, slow-release modifiers, binders and solvents.

One example of a granular composition uses the following constituents:

Example 19

active ingredient	50 g
propylene glycol	25 g
clay (particle size: 0.3 to 0.8 mm)	925 g

As indicated above, the invention also relates to a method of controlling the growth of weeds at a locus used, or to be used for growing crops, in particular cereals, such as wheat, and also soya, which method comprises applying to the locus an effective amount of a compound according to the invention.

The compounds of general formula I may be applied to the plants and/or to the soil. The compounds of the invention are especially suitable for controlling the growth of weeds in a soya crop infested or susceptible to infestation by at least one of the weeds abutilon, xanthium and ipomea, and for controlling the growth of weeds in a cereal crop infested or susceptible to infestation by at least one of the weeds polygonum, chrysanthemum, amaranth, stellaria and galium. In practice, the compounds are used in the form of herbicidal compositions according to the invention, which have been described above. In general, amounts of active ingredient ranging from 0.01 to 5 kg/ha, preferably from 0.1 to 2 kg/ha, give good results, it being understood that the choice of the amount of active ingredient to be used depends on the severity of the problem to be solved, the climatic conditions and the crop in question. The treatment can be carried out either as a pre-emergence treatment of the crops and adventitious plants, or as a pre-sowing treatment of the crops with incorporation into the soil (such incorporation constitutes a further feature of the method of the invention), or as a post-emergence treatment. Other embodiments of the treatment method according to the invention can also be used: thus, it is possible to apply the active ingredient to the soil, with or without incorporation, before planting out a crop.

The treatment method of the invention can be used for annual crops and in the case of perennial crops; in the latter case, it is preferred to apply the active ingredients of the invention in a localised manner, e.g. between the rows of the said crops.

TABLE (I)

American name	*Latin name*	*Abbreviation*
Field corn		CN
Cotton		CT
Wheat		WT
Rice		RI
Soybean		SB
Barnyard grass	Echinochloa crus-galli	BYG
Giant foxtail	Setaria faberii	GTF
Green foxtail	Setaria viridis	GF
Crabgrass	Digitaria sanguinalis	CG
Velvet leaf	Abutilon theophrasti	VL
Foxtail miller	Setaria italica	FM
Cocklebur	Xanthium pennsylvanicum	CB
Wild mustard	Sinapis arvensis	WM
Pigweed	Amaranthus retroflexus	PW
Morning glory (annual)	Ipomea purpurea	MG
Prickly sida	Sida spinosa	PS
Wild oat	Avena fatua	WO
Wild buckwheat	Polygonum convolvulus	
Ragweed	Ambrosia artemisiifolia	

TABLE (II)

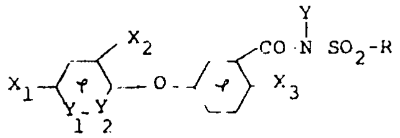
Compound according to Example	Pre-emergence or post-emergence	Dose in kg/ha	Weeds									Crops				
			BYG	GTF	CG	VL	CB	WM	PW	MG	PS	CT	CN	SB	RI	WT
2	pre	1.12	100	90	10	100	--	100	100	100	100	20	40	10		50
		0.56	100	100	100	100	--	100	100	100	100		30	10		
		0.28	100	100	100	100	--	100	100	60	100		30	0		
		0.14	50	60	90	90	--	100	100	20	80	10	20	0	0	20
2	post	1.12	70	70	60	100	100	100	90	80	60	0	0	20	30	30
		0.56	30	70	60	90	100	100	90	80	50	0	0	30	20	30
		0.28	20	50	60	70		100	90	80	20	0	0	10	0	10
		0.14	20	20	20	60	10	100	90	50	20	0	0	10	0	10
1	pre	0.56		0		70	20	90	80	40		20	0	0		10
1	post	0.56		10		30	20	90	20	10		30	10	20		0

TABLE (III)

Dose in kg/ha	PRE-EMERGENCE: POST-EMERGENCE						
	1	0.5	0.25	1	0.5	0.25	0.125
Agropyron	90	60	0				
Sorghum	100	100	100				
Echinochloa	100	100	100	100	90	60	30
Panicum	100	100	100	100	100	100	100
Digitaria	100	100	100	100	100	90	80
Abutilon	100	100	100	100	100	100	100
Xanthium	100			100	100	100	100
Chenopodium	100	100	100	80	80	80	80
Amaranthus	100	100	100		100	100	100
Ambrosia	100	100	100	100	100	100	100
Polygonum	100	100	100	100	100	100	100
Ipomea	100	100	50	100	100	90	90
Sida	100	100	100	100	100	100	100
Setaria viridis	100	100	100	90	90	80	10
Alopecurus	80	80	10				
Lolium	100	80	60				
Setaria Faberii	100	100	100	80	20	20	0
Chrysanthemum	100	100	100	100	100	100	100
Sinapis	100	100	100	100	100	100	100
Stellaria	100	100	100				
Galium	100	100	100	90	90	90	10
Wheat	80	60	10	10	10	0	0
Barley	20	20	10	10	0	0	0
Maize				50	20	10	0
Cotton	-	0	0				
Soya	0	20		0	0	0	0
Cyperus	80		0	60	20	0	0

CLAIMS

1. A compound of the formula:



(I)

in which: Y_1 is a nitrogen atom or a group $-CH=$, Y_2 is a nitrogen atom or, when Y_1 is a group $-CH=$, a group $-CX_4=$, X_1, X_2 and X_3, which are identical or different, each represent a halogen atom or a polyhalogenoalkyl, cyano, nitro, alkyl, alkoxy, alkylsulphonyl, $-SO_2NH_2$, nitroso or alkoxycarbonyl group, X_4 is as defined for X_1, X_2 and X_3 and can also represent hydrogen, R is a substituted or unsubstituted hydrocarbon radical or a substituted or unsubstituted heterocyclic ring and Y is a hydroxy group or a halogen atom.

2. A compound according to claim 1 in which alkyl groups and moieties within the definition of X_1 to X_4 contain from 1 to 4 carbon atoms, and in which R is a substituted or unsubstituted hydrocarbon radical having from 1 to 12 carbon atoms or a substituted or unsubstituted heterocyclic ring having from 5 to 7 atoms in the ring.

3. A compound according to claim 1 or 2, in which R is an alkyl radical having from 1 to 4 carbon atoms, a phenyl radical which is unsubstituted or substituted by 1 to 4 alkyl radicals each having from 1 to 4 carbon atoms, by chlorine or bromine atoms, by hydroxy, SH, CN or COOH groups or by alkoxy, thioalkyl or alkoxy-carbonyl groups, the alkyl parts of these groups having from 1 to 4 carbon atoms or an alkenyl or alkynyl group having from 2 to 6 carbon atoms.

4. A compound according to claim 1, 2 or 3 in which X_1 and X_2 are polyhalogenoalkyl groups or halogen atoms, Y_1 and Y_2 both represent the group $-CH=$ and X_3 is the nitro group or a halogen atom.

5. A compound according to any one of claims 1 to 4 in which X_1 is Cl or CF_3, X_2 is Cl, X_3 is Cl or NO_2 and Y_1 and Y_2 are both $-CH=$.

6. A compound according to claim 5 in which X_1 is CF_3 and X_3 is NO_2.

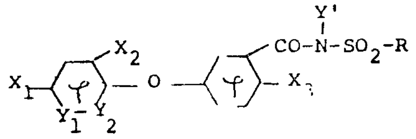
7. A compound according to any one of the preceding claims in which Y is a chlorine atom.

8. A compound according to any one of the preceding claims in which R is an alkyl group of 1 to 4 carbon atoms and Y is a chlorine atom.

9. 5-(2,4-Dichlorophenoxy)-2-nitro-N-chloro-N-methanesulphonylbenzamide.

10. 5-[2-Chloro-4-(trifluoromethyl)-phenoxy]-2-nitro-N-chloro-N-methanesulphonylbenzamide.

11. A process for the preparation of a compound of general formula I as defined in claim 1 wherein Y is a halogen atom, which comprises reacting a hypohalite with a compound of the formula:



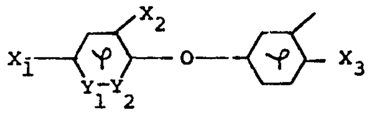
(III)

in which Y' is a hydrogen atom or an alkali metal atom and the other symbols are as defined in claim 1.

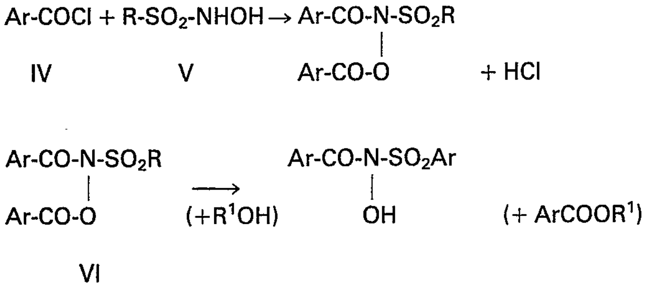
12. A process according to claim 11, wherein the reaction is carried out in an aqueous medium at from −10 to +60°C, the concentration of reactant of the formula (III) and reaction product of the formula (I) in the reaction medium being from 0.5 to 30% by weight, and the pH being more than 10.

13. A process according to claim 11 or 12, wherein the temperature is from 10 to 40°C, the concentration is from 2 to 10% and the pH is more than 11.5.

14. A process for the preparation of a compound of general formula I as defined in claim 1 wherein Y is a hydroxy group which comprises reacting a compound of formula IV with a compound of formula V to obtain a compound of formula VI and then reacting the compound of formula VI with an alkali metal alkoxide or with a solution of an alkali metal hydroxide in an alcohol R^1OH in which R^1 represents an alkyl group and then with a strong acid to obtain the compound of formula I in which Y is a hydroxy group, in accordance with the following reaction scheme in which the symbol Ar represents the radical:



R′ represents an alkyl group and the other symbols are as hereinbefore defined:



$$\underset{IV}{\text{Ar-COCl}} + \underset{V}{\text{R-SO}_2\text{-NHOH}} \rightarrow \begin{array}{c}\text{Ar-CO-N-SO}_2\text{R}\\ |\\ \text{Ar-CO-O}\end{array} + \text{HCl}$$

$$\underset{VI}{\begin{array}{c}\text{Ar-CO-N-SO}_2\text{R}\\ |\\ \text{Ar-CO-O}\end{array}} \xrightarrow[(+R^1OH)]{} \begin{array}{c}\text{Ar-CO-N-SO}_2\text{Ar}\\ |\\ \text{OH}\end{array} \quad (+ \text{ArCOOR}^1)$$

15. A process according to any one of claims 11 to 14 substantially as hereinbefore described.

16. A process according to claim 11 substantially as hereinbefore described in Example 1 or 2.

17. A compound according to claim 1 when prepared by a process according to any one of claims 11 to 16.

18. A herbicidal composition which comprises, as active ingredient, a compound according to any one of claims 1 to 10 and 17 in association with a herbicidally acceptable carrier.

19. A composition according to claim 18, which contains 0.05 to 95% by weight of active ingredient.

20. A composition according to claim 18 or 19, which is liquid and which contains 10 to 80% of active ingredient.

21. A composition according to claim 18 or 19, which is solid and contains 20 to 95% of active ingredient.

22. A composition according to any one of claims 18 to 21, which contains 0.1 to 20% of surface-active agent.

23. A composition according to claim 18 substantially as hereinbefore described in any one of Examples 4 to 19.

24. A method of controlling the growth of weeds at a locus used, or to be used for growing crops, which comprises applying to the locus an effective amount of a compound according to any one of claims 1 to 10 and 17.

25. A method according to claim 24, wherein the composition is applied to a soya crop infested or susceptible to infestation by at least one of the weeds abutilon, xanthium and ipomea.

26. A method according to claim 24 wherein the crop is a cereal crop infested or susceptible to infestation by at least one of the weeds polygonum, chrysanthemum, amaranth, stellaria and galium.

27. A method according to any one of claims 24 to 26 wherein the compound of the formula (I) is applied at a rate of 0.01 to 5 kg/ha.

28. A method according to any one of claims 24 to 26, wherein the compound of the formula (I) is applied at a rate of 0.1 to 2 kg/ha.

29. A method according to claim 24 substantially as hereinbefore described.

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